

# Exploring atomic dynamics under extreme conditions

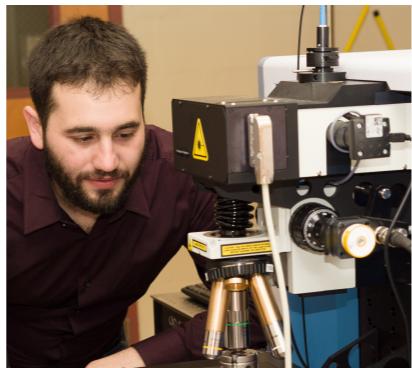
Jennifer M. Jackson  
California Institute of Technology



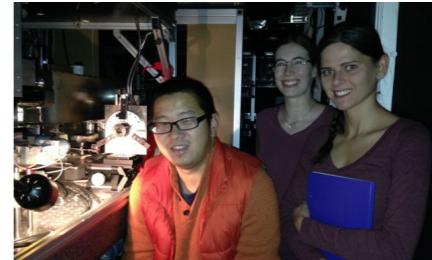
13 July 2015

Emerging Opportunities in High Energy X-ray Science: The Diffraction  
Limited Storage Ring Frontier  
SRI Satellite Workshop, Advanced Photon Source

# Acknowledgements



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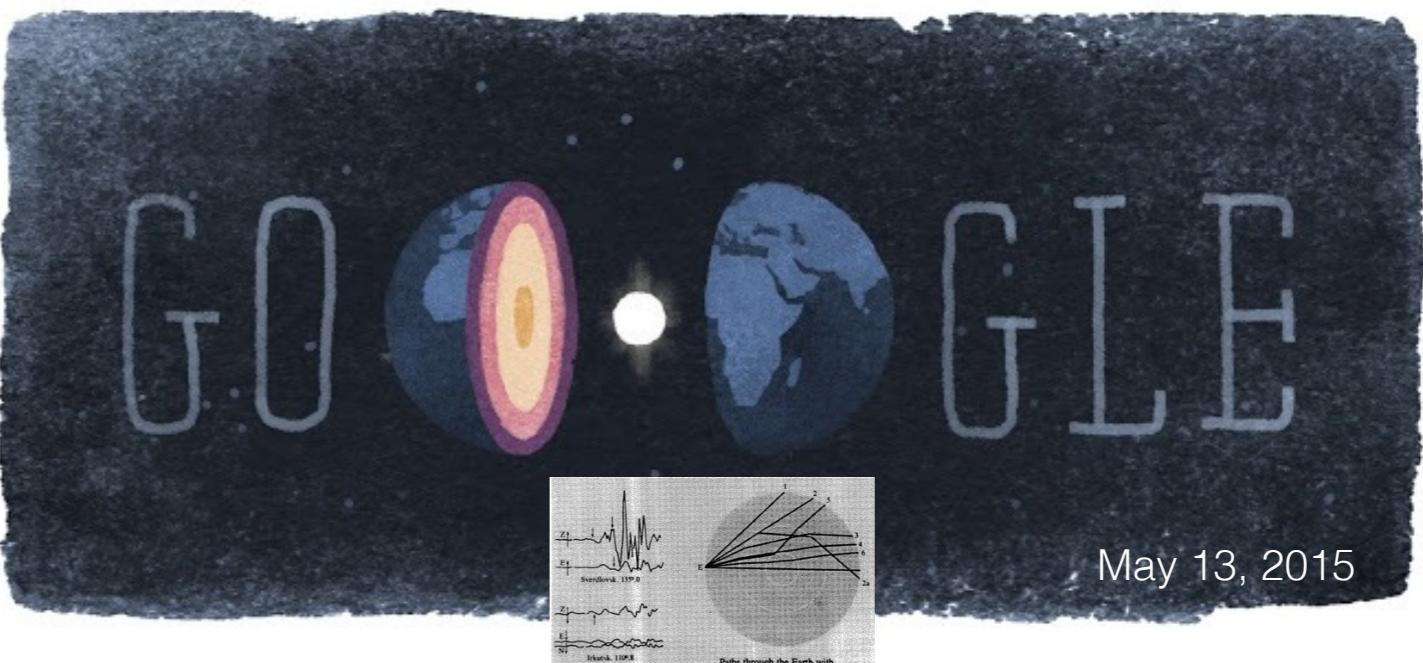


CSEDI  
CAREER  
Geophysics

# Probes have improved models of Earth's and planetary interiors



Athanasius Kircher (1665)  
Mundus Subterraneus



May 13, 2015

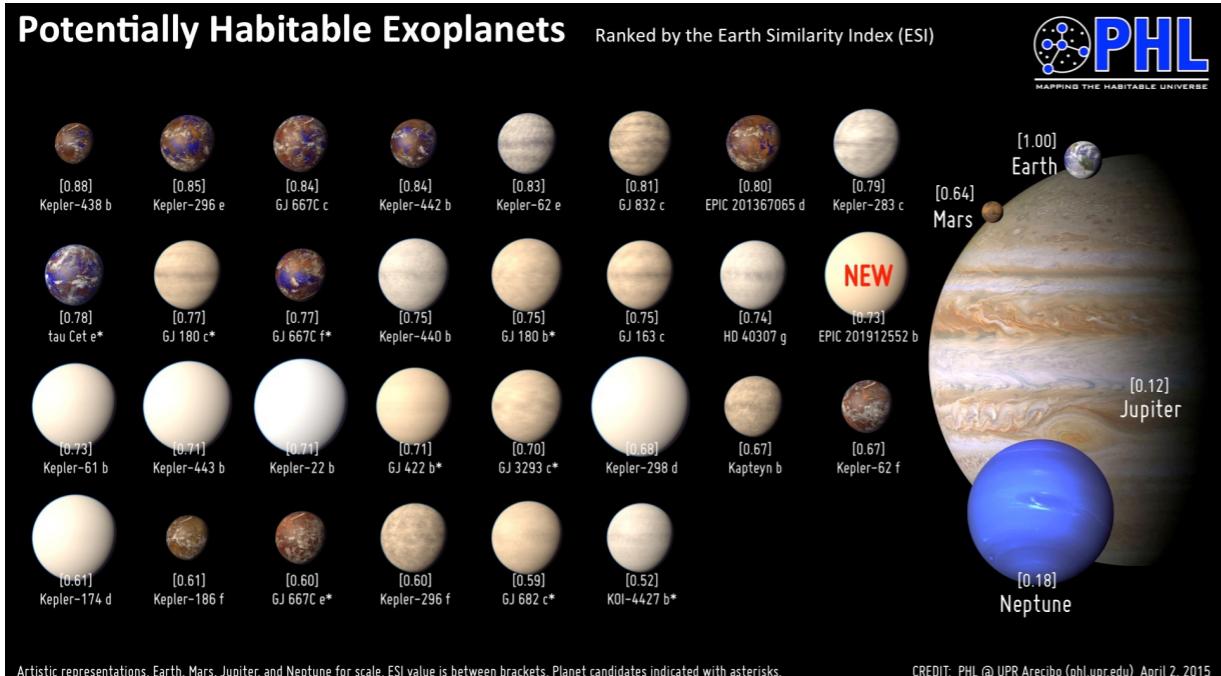
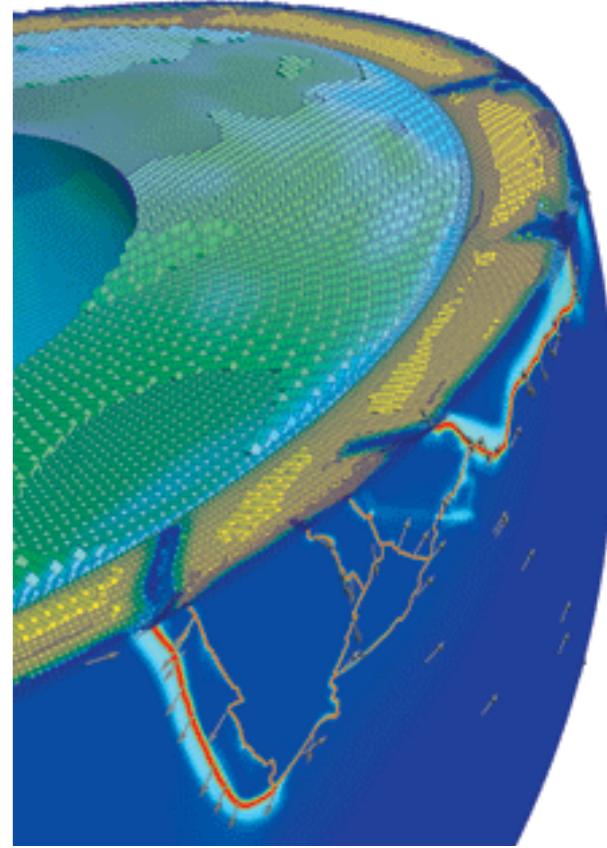
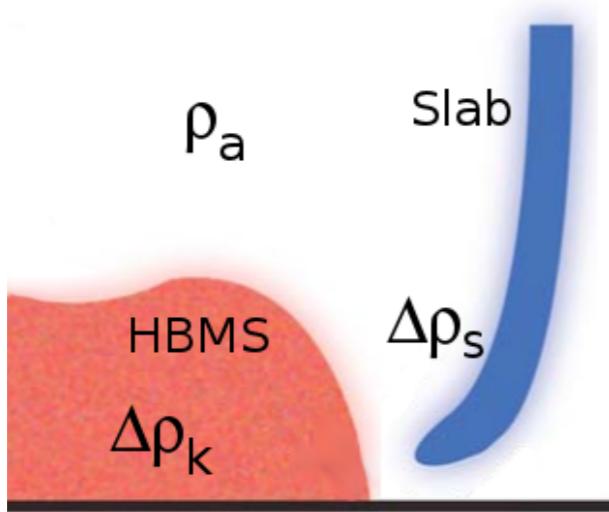


Image credit: Planetary Habitability Laboratory  
Univ. of Puerto Rico, Arecibo ([phl.upr.edu](http://phl.upr.edu))

seismology  
gravity and magnetic fields  
cosmochemical models  
material properties  
geodynamical modeling  
long-period librations  
mass/radius relationships

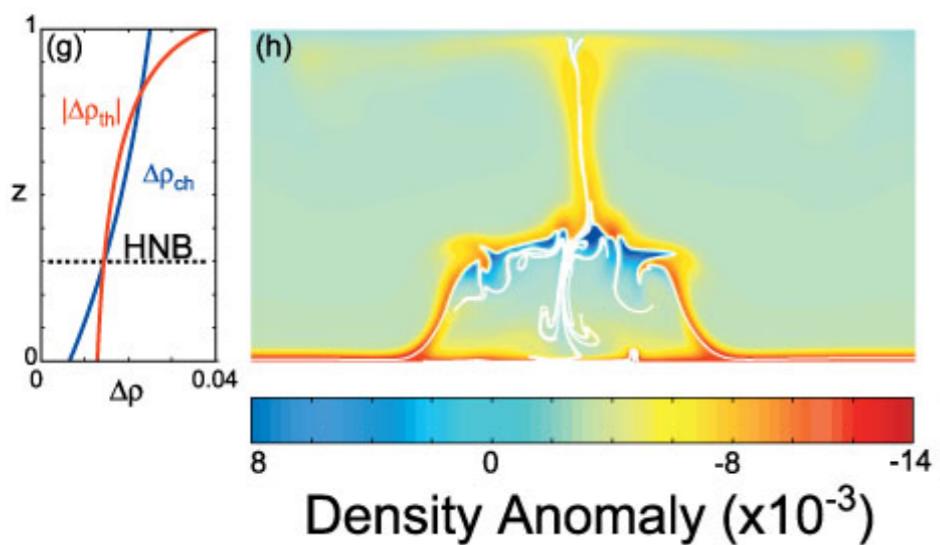


# Large coherent structures resting on Earth's core-mantle boundary: composition? lifetimes?



## Passive Piles

More dense than surrounding mantle, dynamically swept upward into piles  
adapted from Tan *et al.* *G3* (2011).  
e.g., Tackley 2011, Li & McNamara 2013, Bower *et al.* *G3* 2013



## Metastable Domes

Less dense than surrounding mantle  
e.g., Tan & Gurnis *GRL* 2005, *JGR* 2007, Sun *et al.* *PNAS* 2007

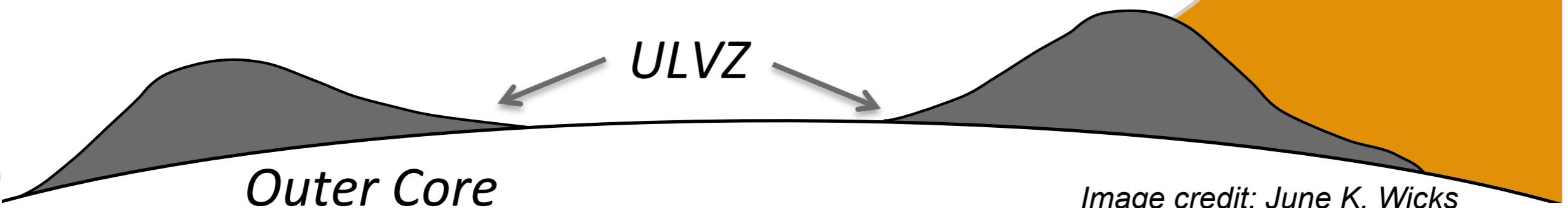
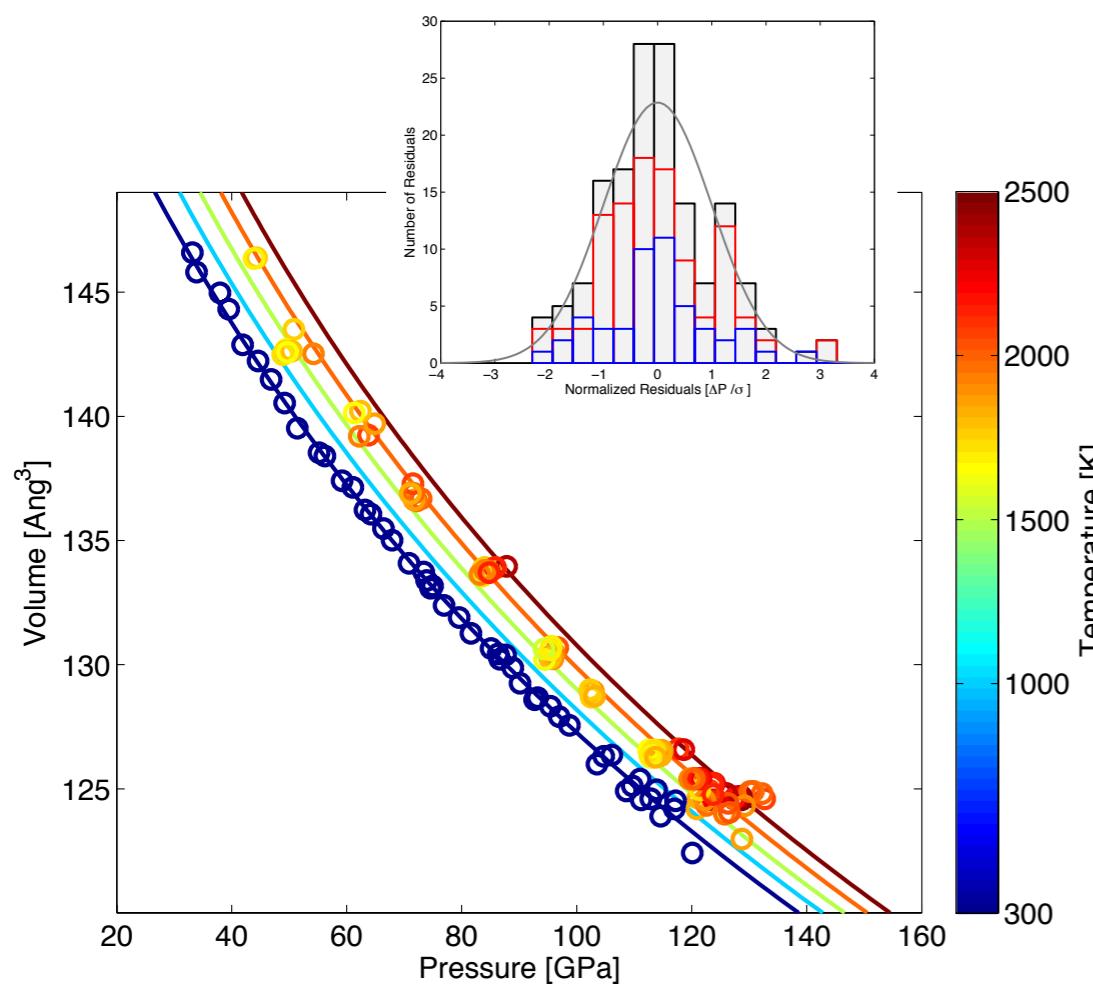
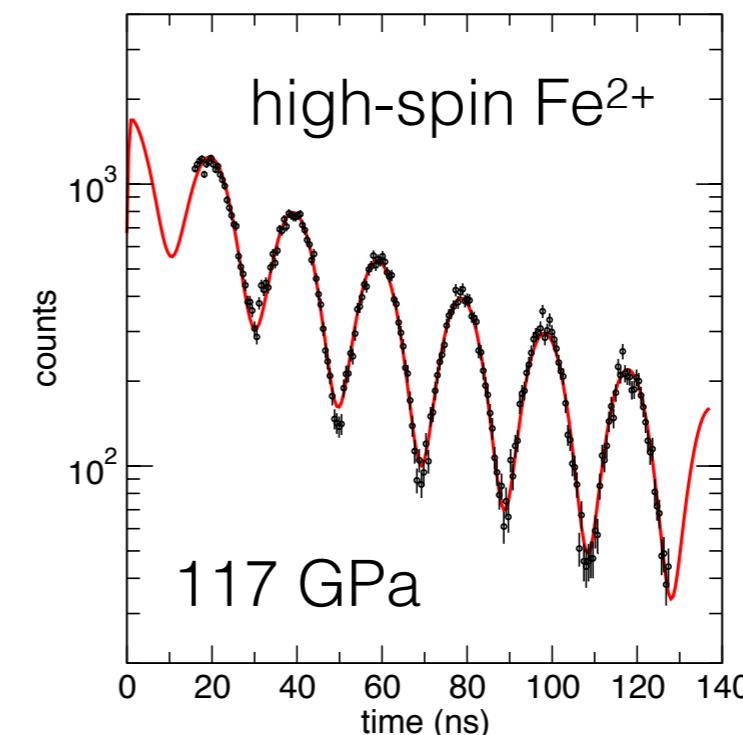
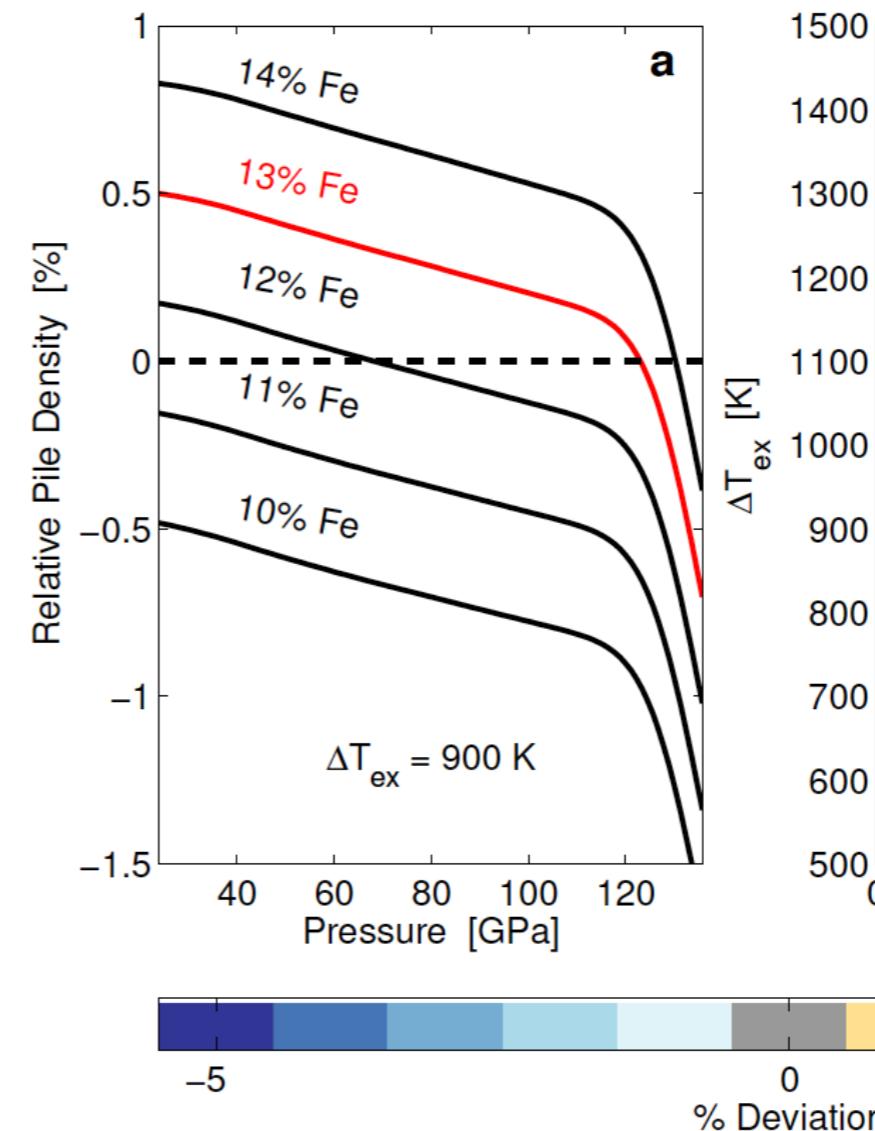


Image credit: June K. Wicks

# Iron-bearing bridgmanite piles?



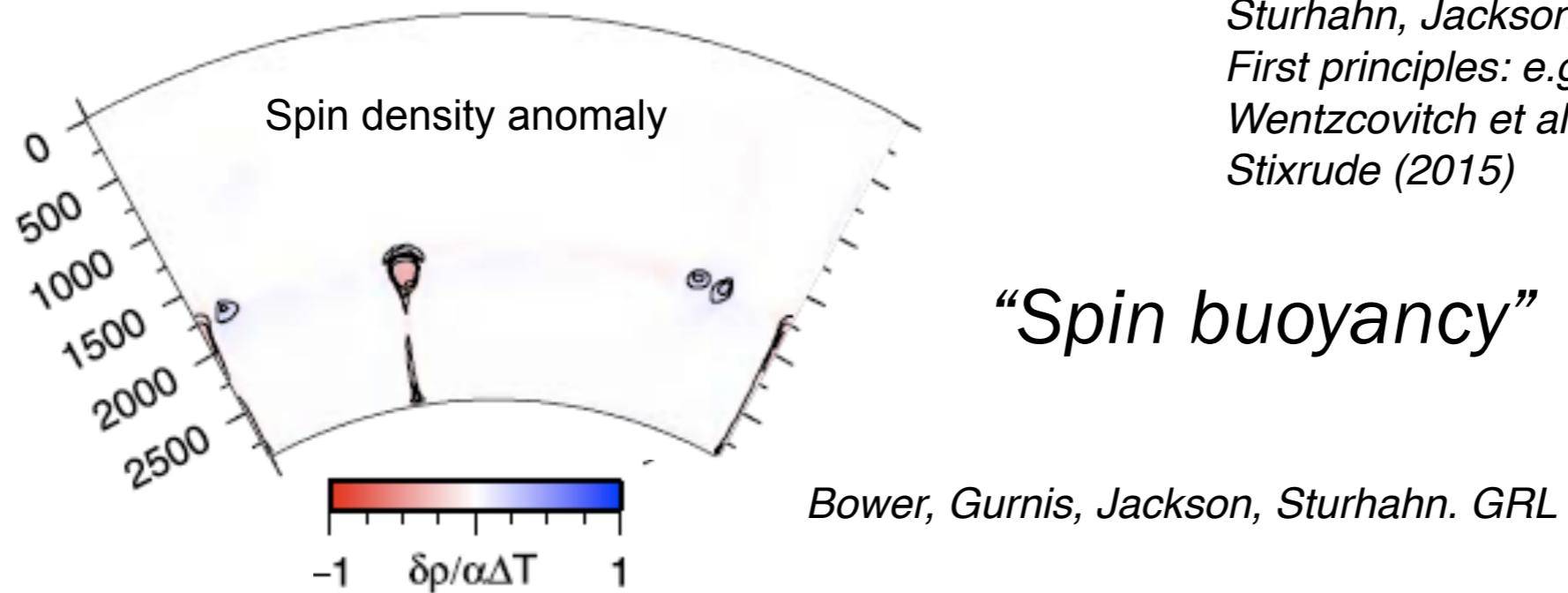
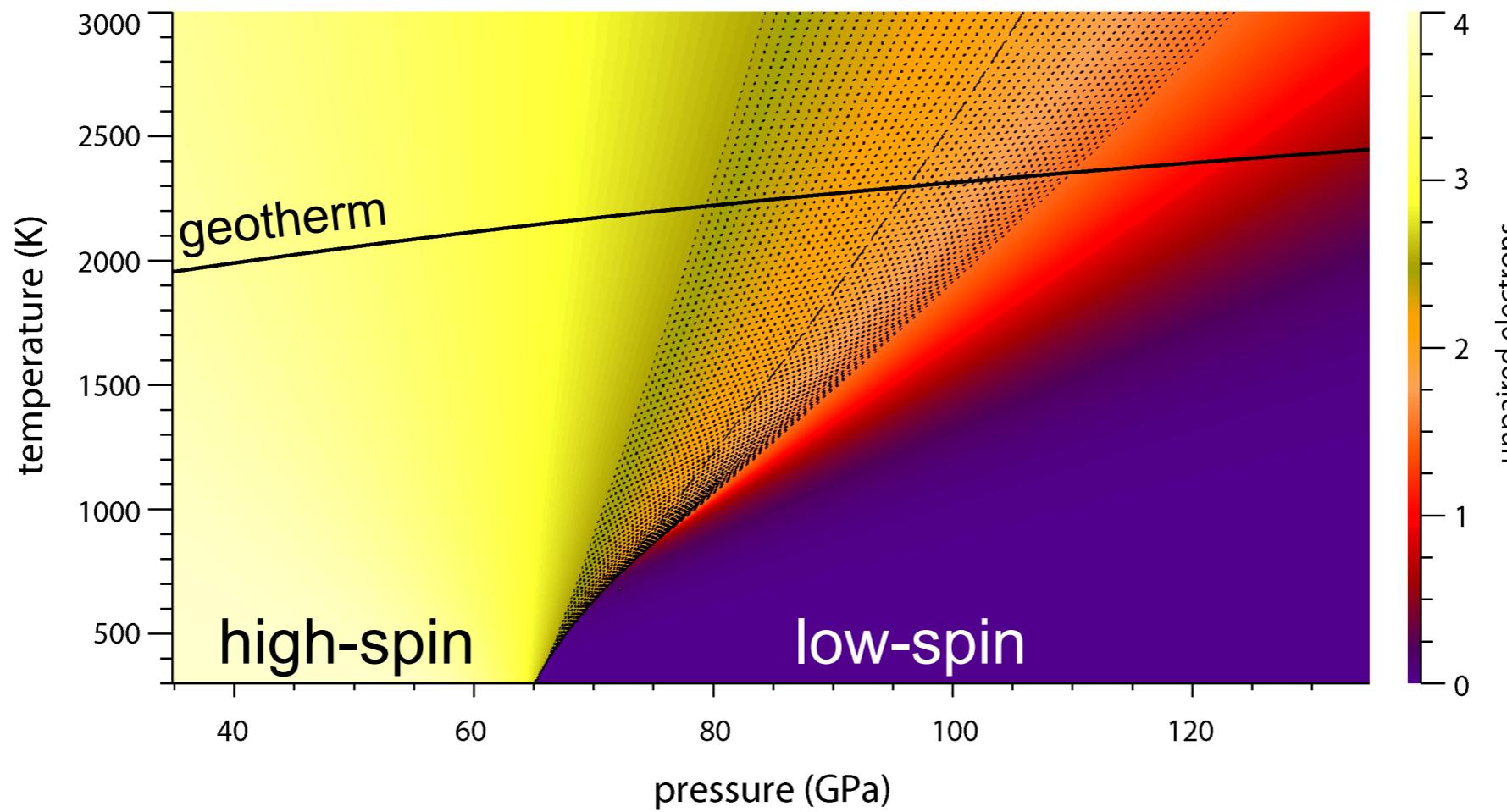
Wolf, Jackson, Dera, Prakapenka  
(2015, under revision)



*in-situ*  
synchrotron Mössbauer  
spectroscopy (SMS):  
valence, spin, magnetism

  
[www.nrixs.com](http://www.nrixs.com)

# Influence of the continuous spin crossover on mantle dynamics



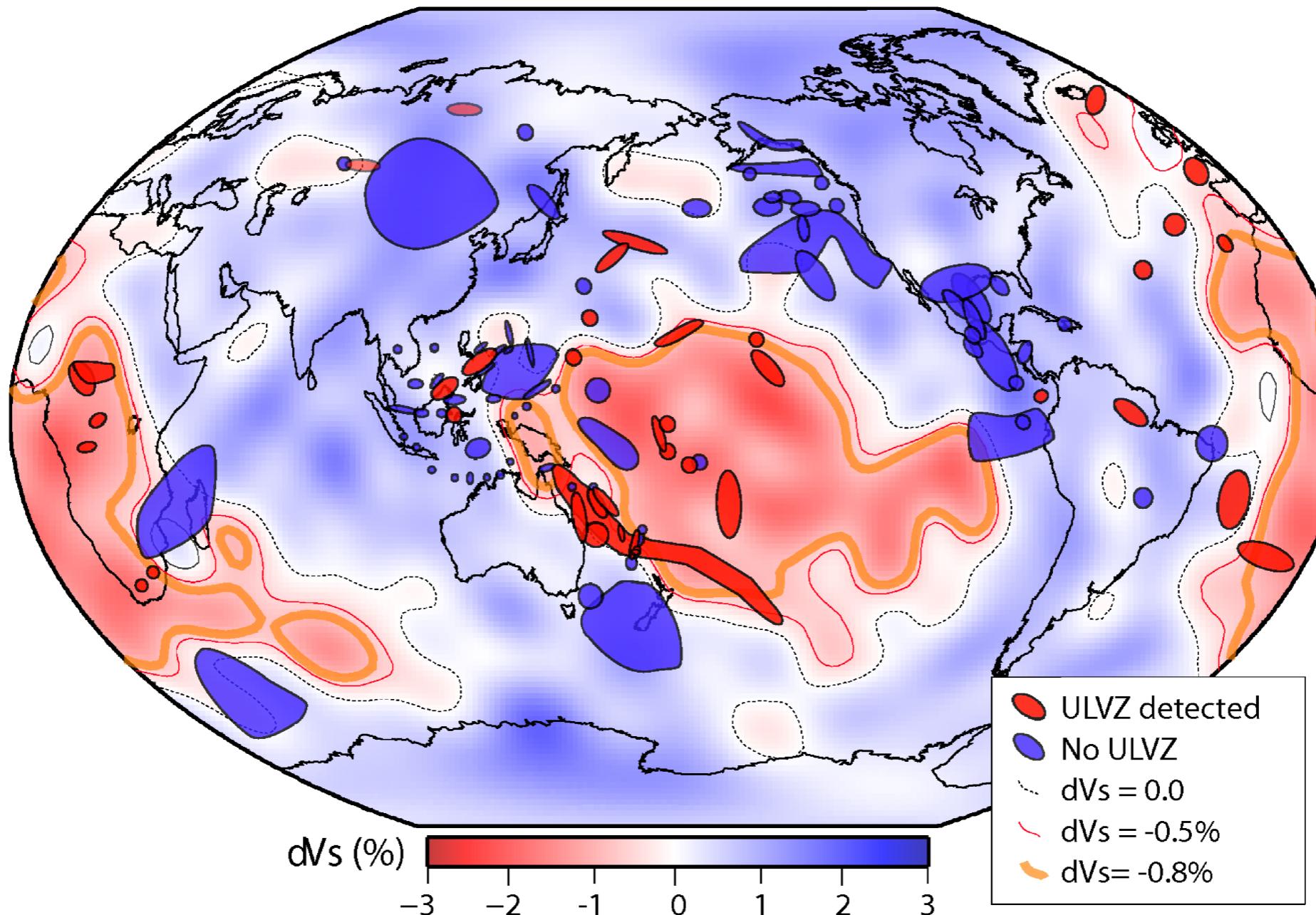
*Sturhahn, Jackson, Lin., GRL (2005)  
First principles: e.g., Tsuchiya et al. (2006),  
Wentzcovitch et al. (2009); Holström &  
Stixrude (2015)*

*Bower, Gurnis, Jackson, Sturhahn. GRL (2009)*

Advectional heat transport more effective

# Distribution of low velocity zones at the base of the mantle

(McNamara et al. *EPSL* 2010)



Slow

~10 to 30% drop in  $V_S$   
~5 to 20% drop in  $V_P$

Dense

~4 to 14% denser

Variable

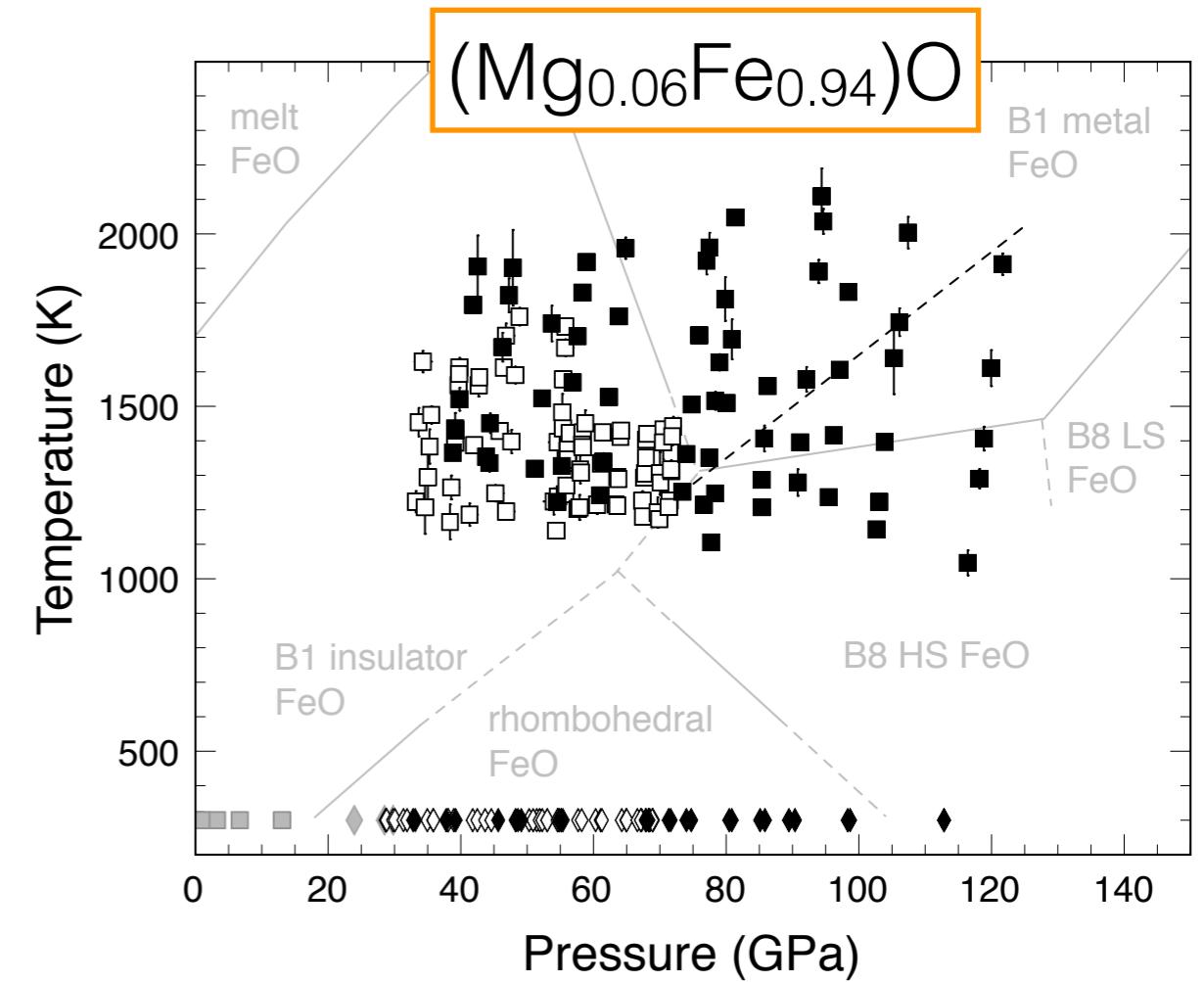
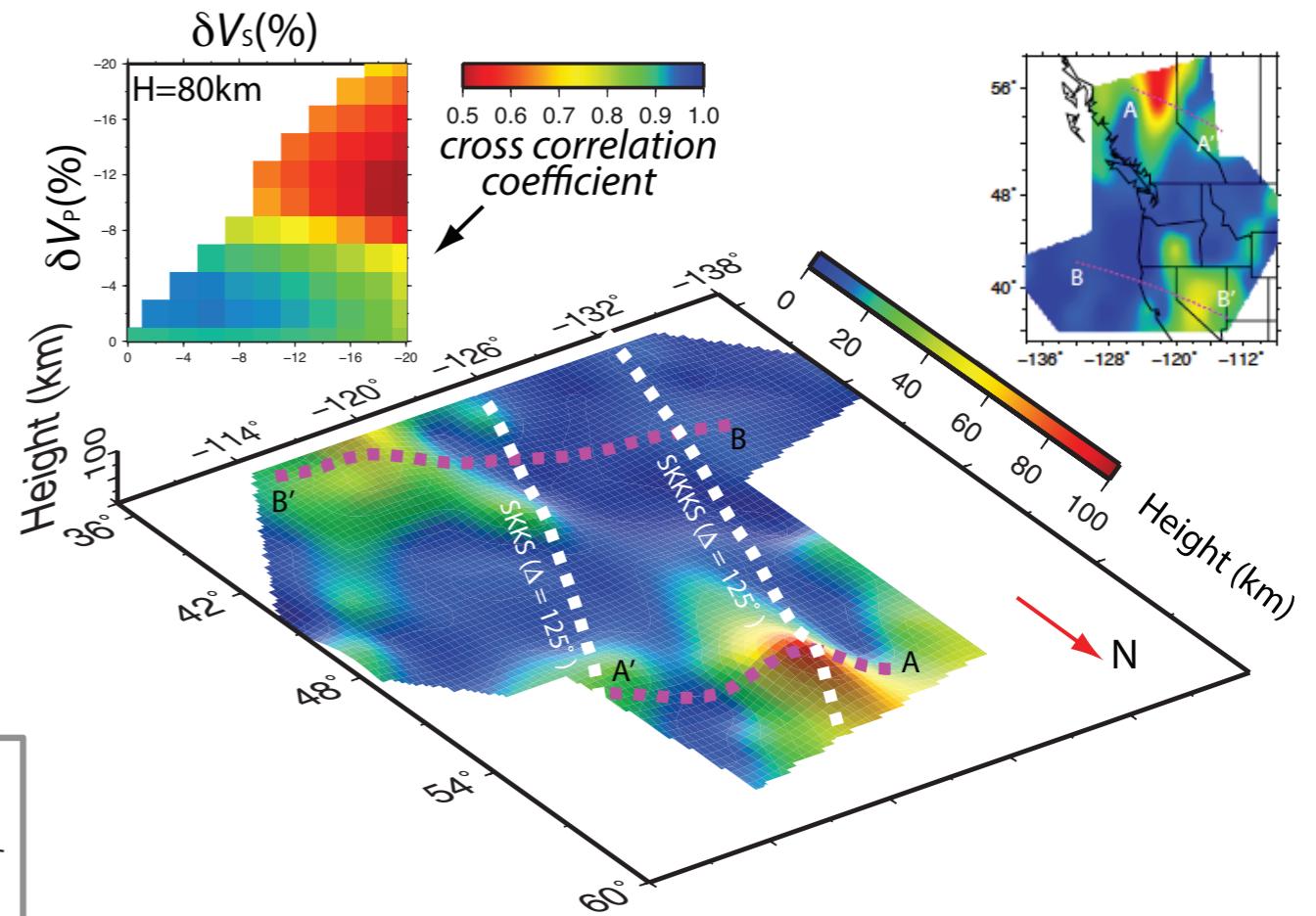
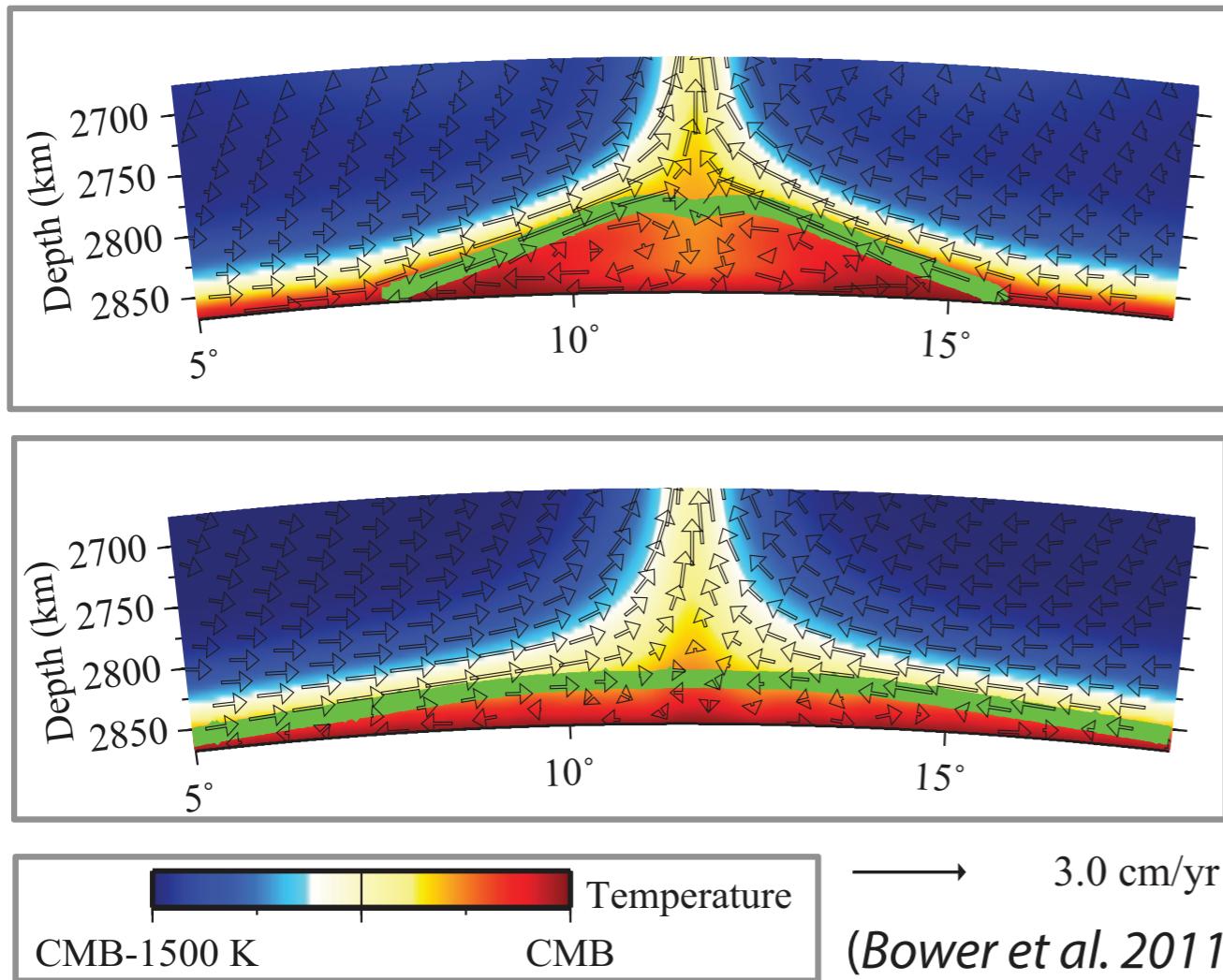
$V_P/V_S$  range ~1 to 5

*Not well-constrained*



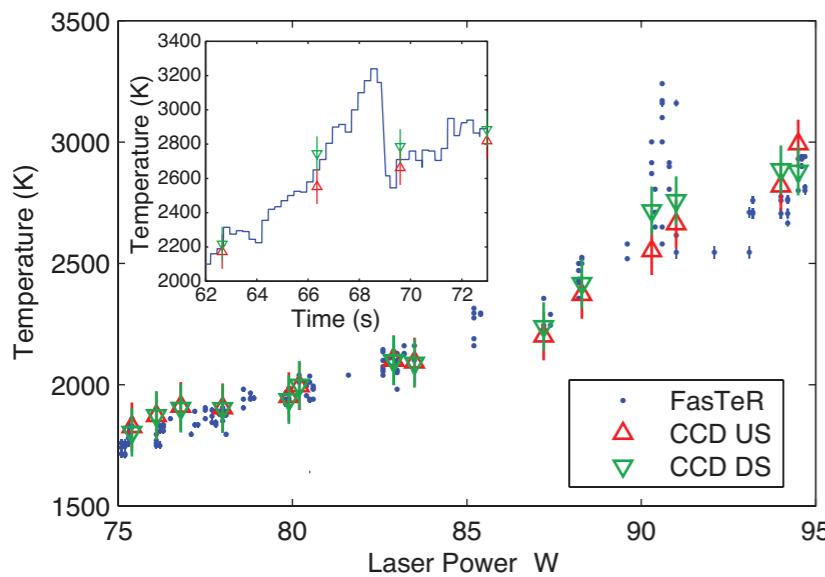
# Dynamics of hot, dense materials at the core-mantle boundary

Wicks et al. GRL 2010, Bower et al. EPSL 2011, Sun et al. EPSL 2013  
 Wicks et al. 2015, under revision

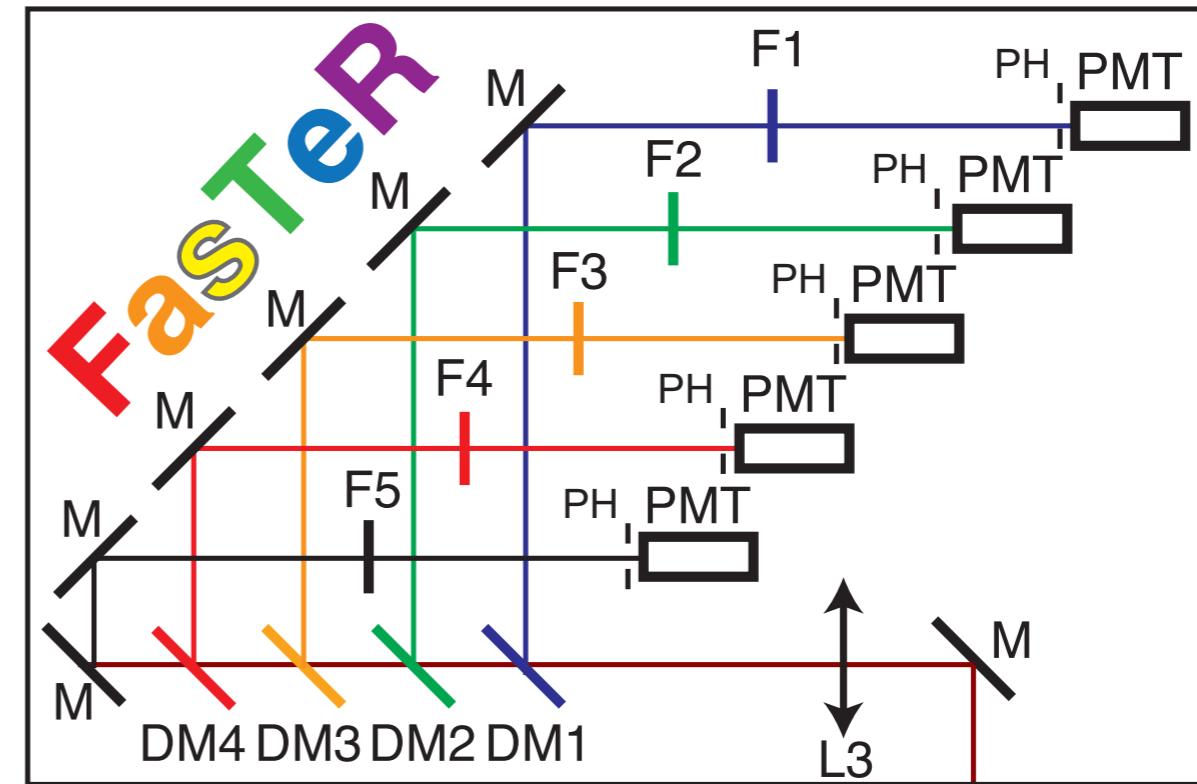
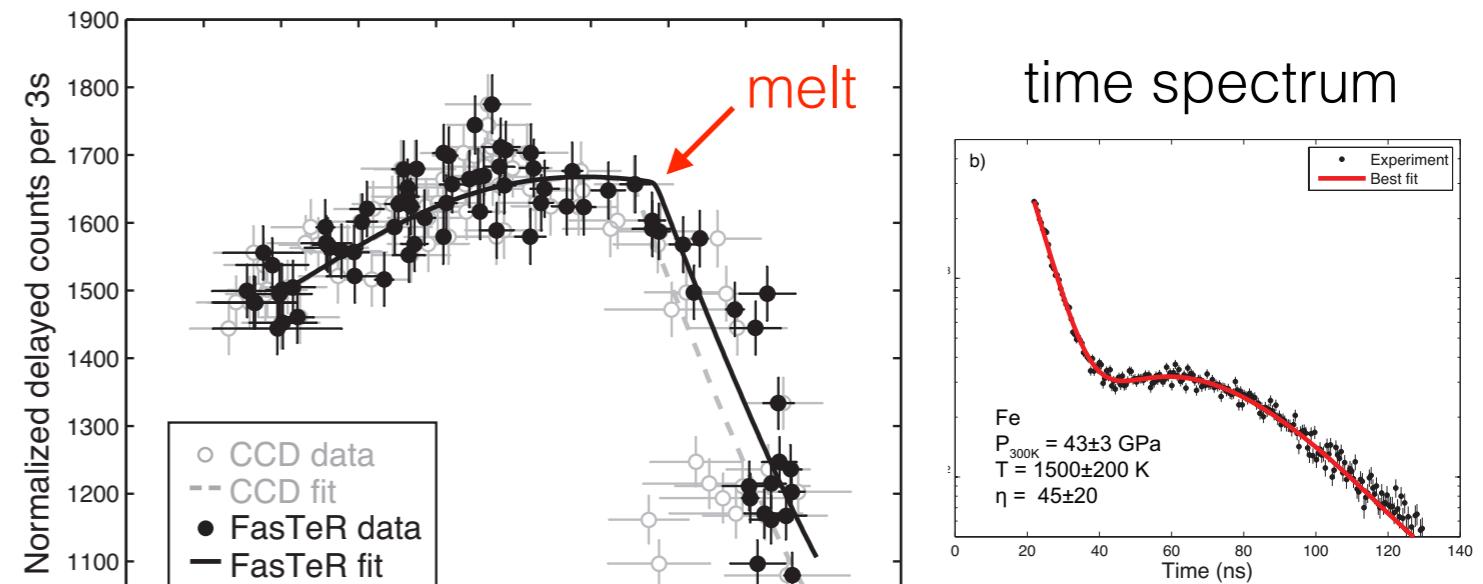


# New FasTeR spectrometer for Mössbauer melting and ultra-fast scattering measurements

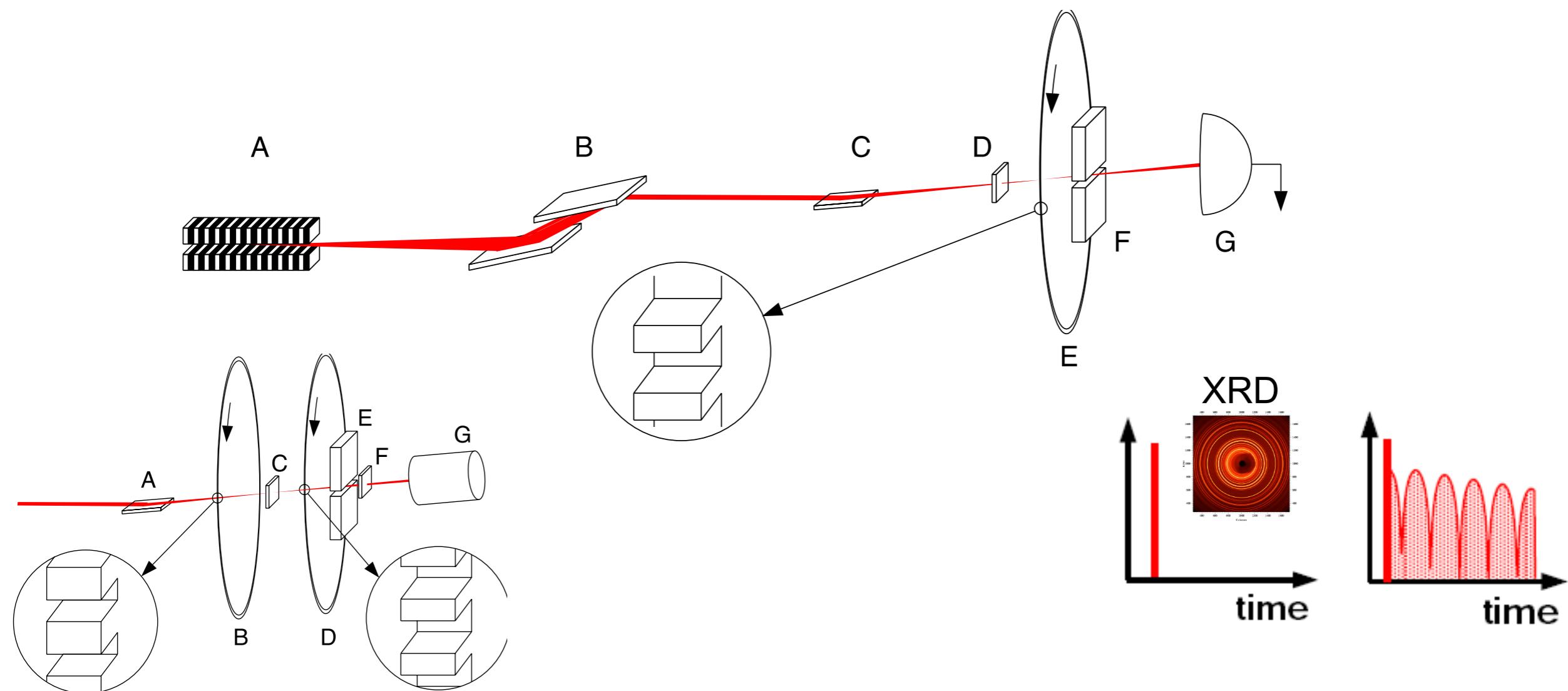
- Fast Temperature Readout system (FasTeR): microsecond readout, up to 400 Hz, combined with synchrotron Mössbauer spectroscopy.
- Target applications: monitoring fast atomic dynamics at high temperatures with high precision



Mössbauer *Z. Phys.* (1958),  
Singwi & Sjölander *Phys. Rev.*  
(1960), Boyle *et al. Proc. Phys.*  
*Soc.* (1960), Jackson *et al. EPSL*  
(2013), Zhang *et al. RSI* (2015)



# Next Generation Mössbauer Spectroscopy With High-Speed Shutters: 4D imaging of Fe valences with diffraction

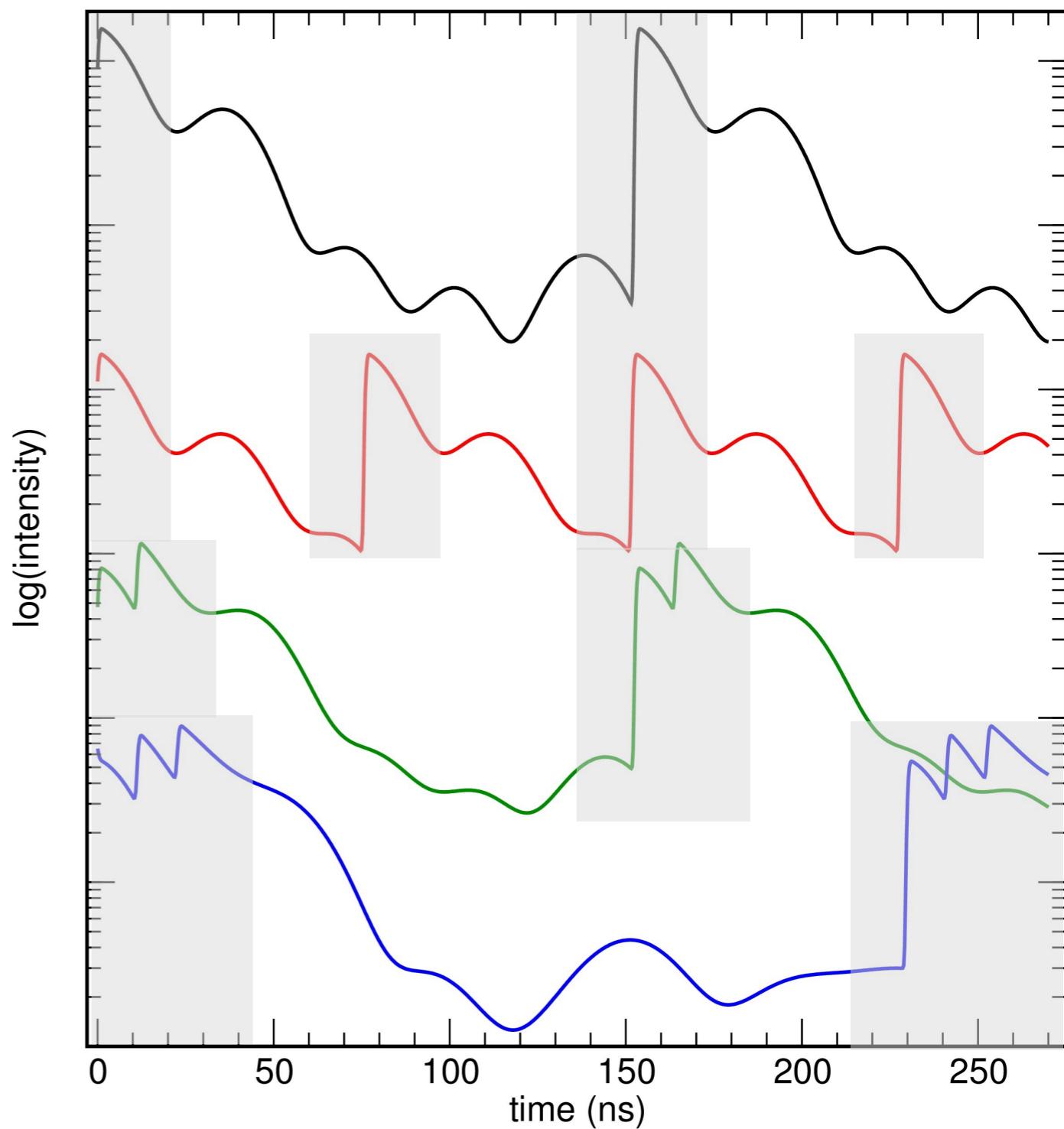
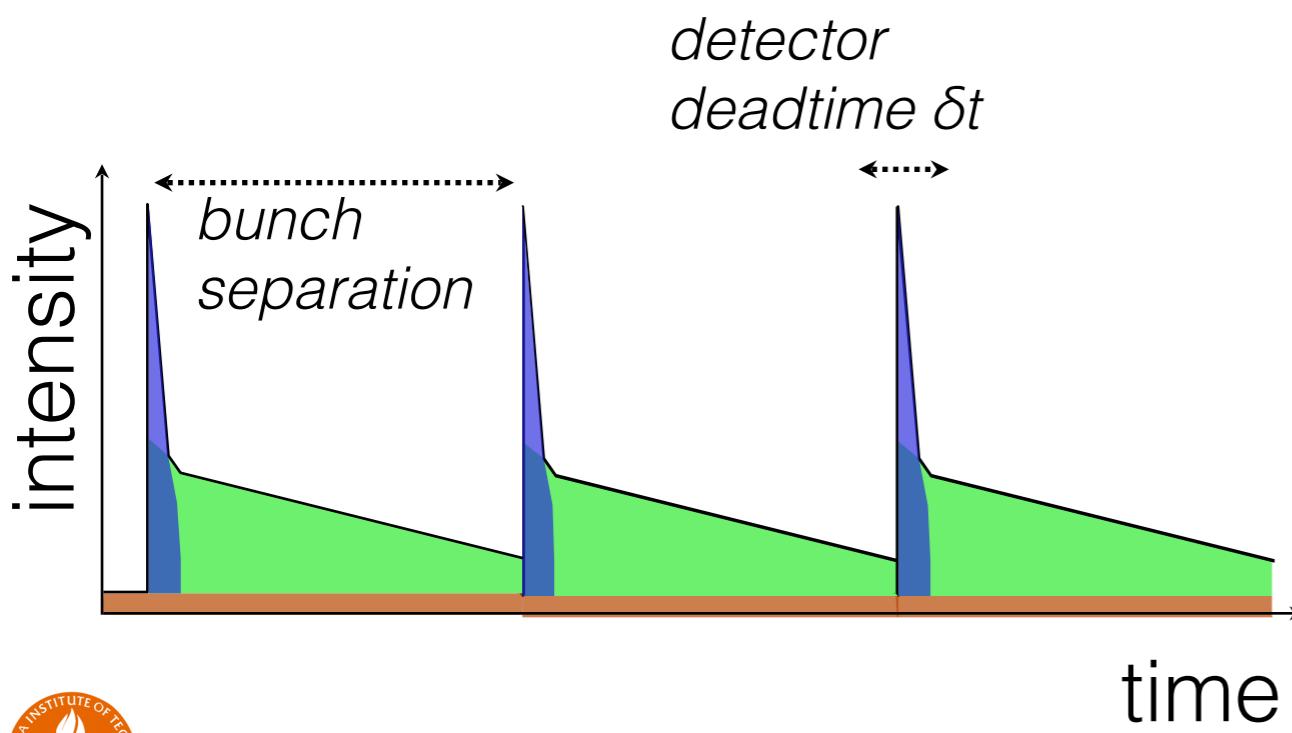


Schematic of high-speed shutters at sector 3, Advanced Photon Source  
(e.g., Toellner *et al.* *JSR* 2010)

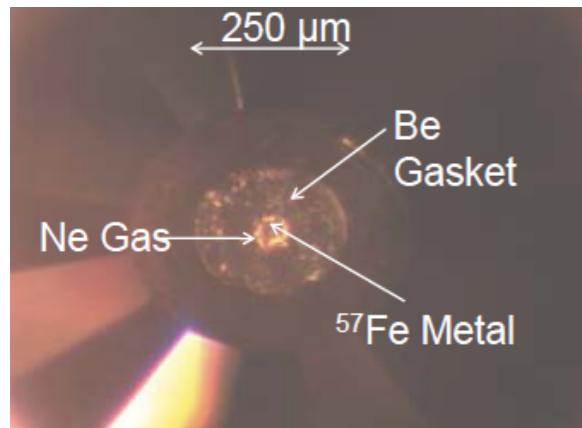
Great advantage for studies of melting  
and other properties related to atomic dynamics (transport)

# MBA for time-differential studies

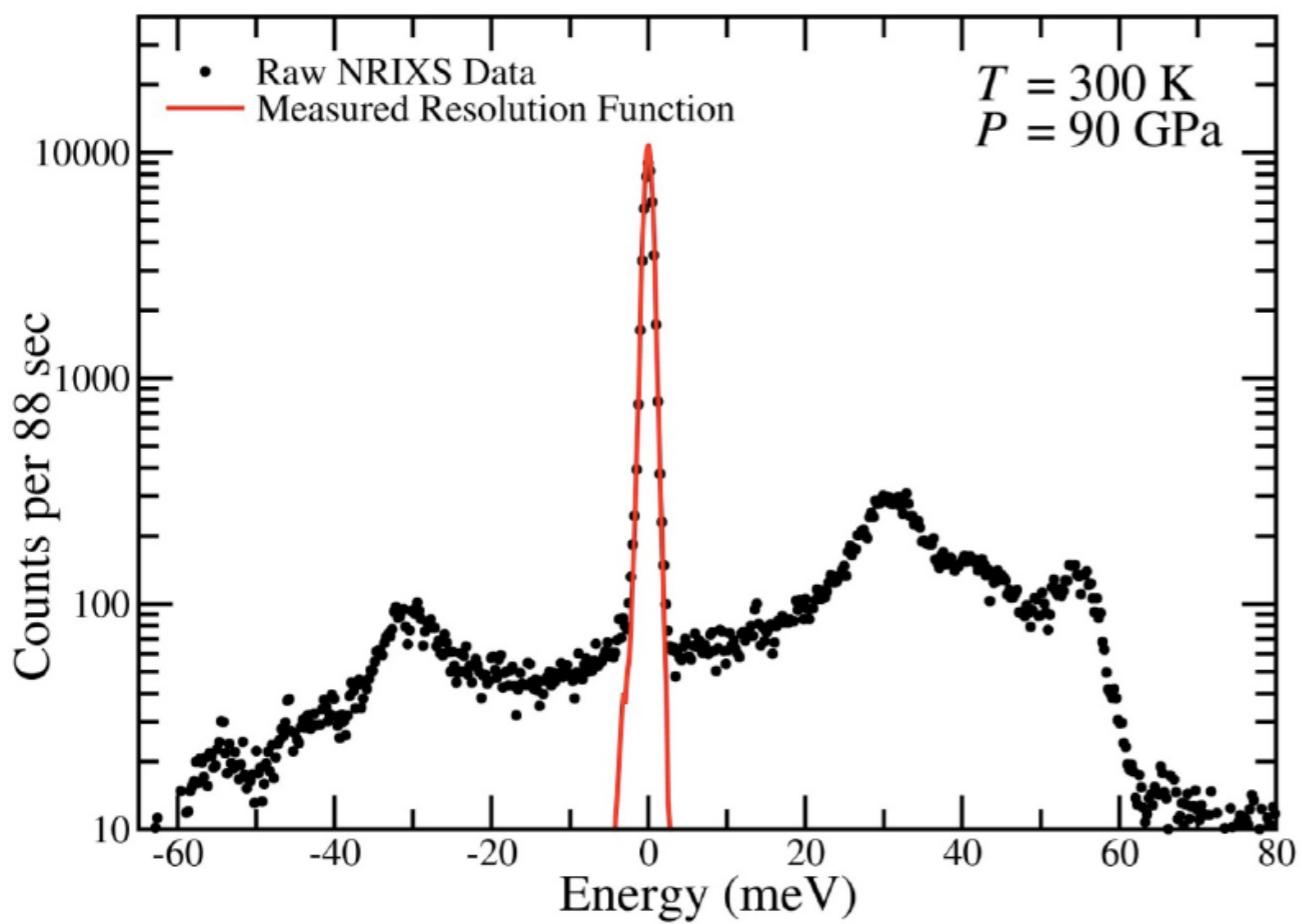
- Figure: time-differential intensity for SMS studies under extreme conditions; grey boxes indicate time regions inaccessible to experiment
- potential MBA timing modes:
  - red, 48-27-1 (singlet mode)
  - green, 24-54-2 (doublet mode)
  - blue, 16-81-3 (triplet mode)
- present day timing mode:
  - black, 24-54-1 (singlet mode)
- MBA timing modes 24-54-2 and 16-81-3 maintain a sufficiently long time span for data collection at multi-Mbar pressures.



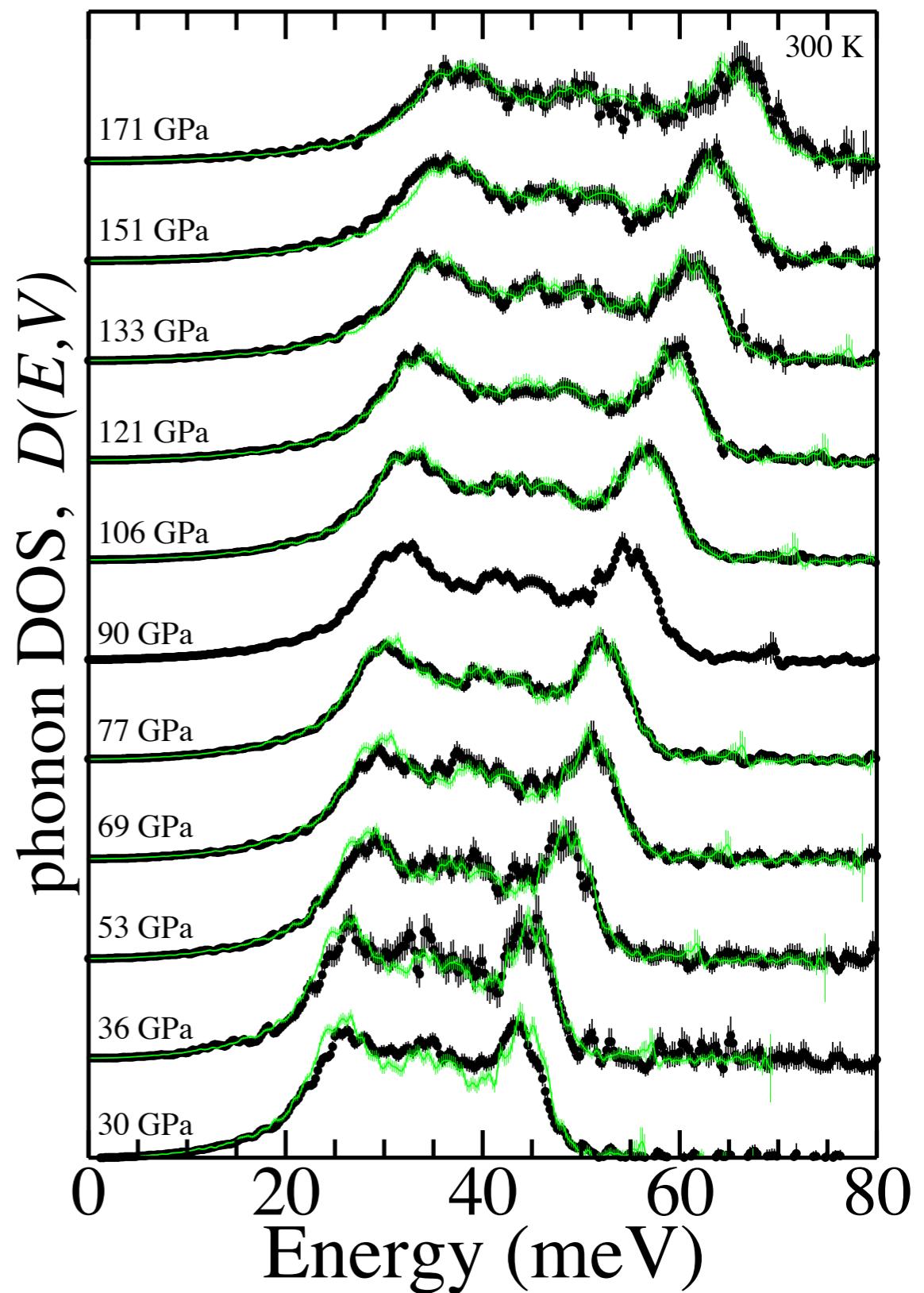
calculations by:  
Wolfgang Sturhahn (Caltech)



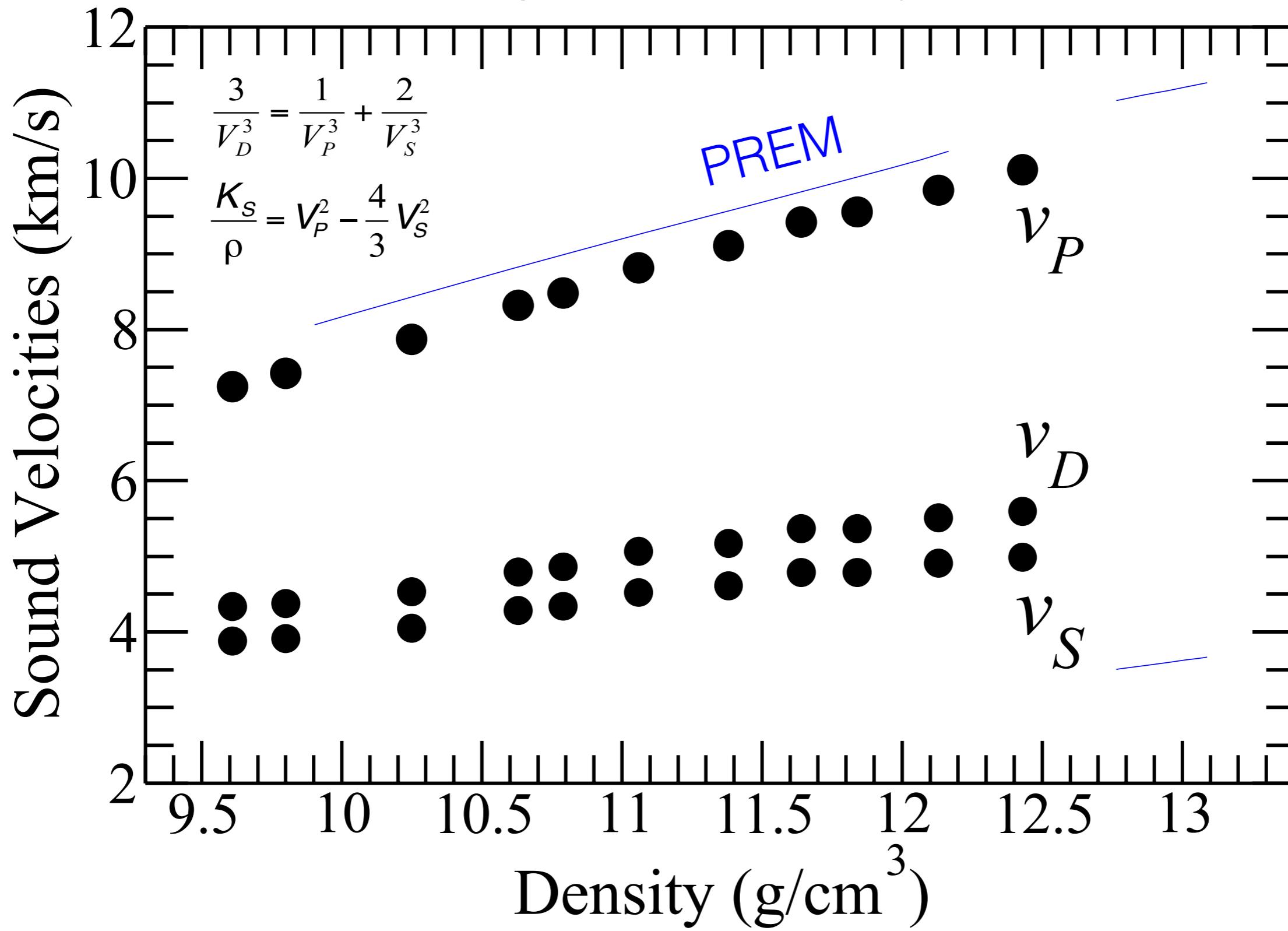
# Volume dependence of the phonon DOS for $hcp$ -iron to outer core pressures



Fe sample loaded with neon  
Pressure scale: Dewaele *et al.* *PRL* (2006)  
Murphy *et al.* *GRL* (2011); Murphy *et al.* *PEPI* (2011);  
Murphy *et al.* *JGR* (2013)



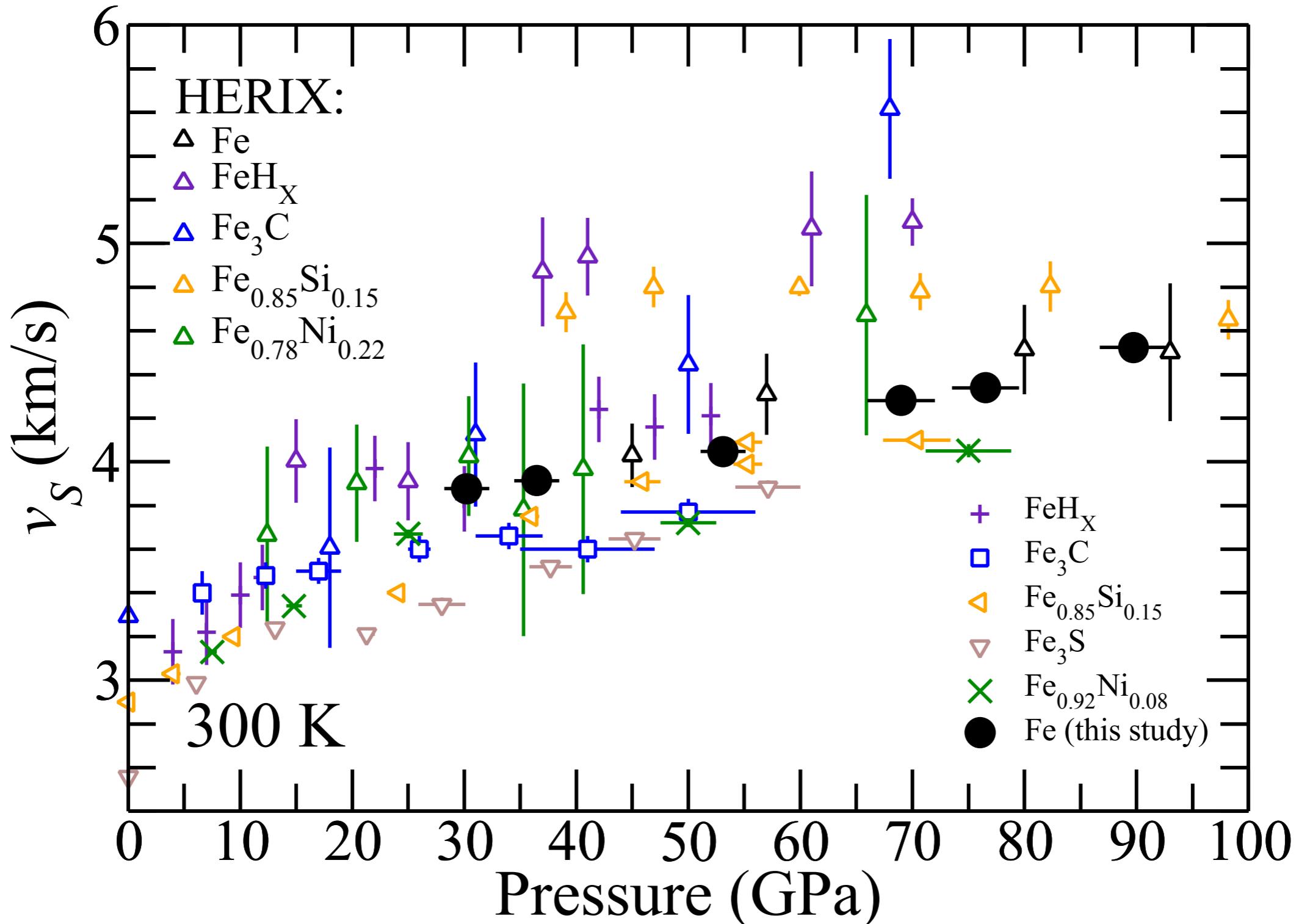
# Sound velocities of *hcp*-iron from the measured phonon density of states



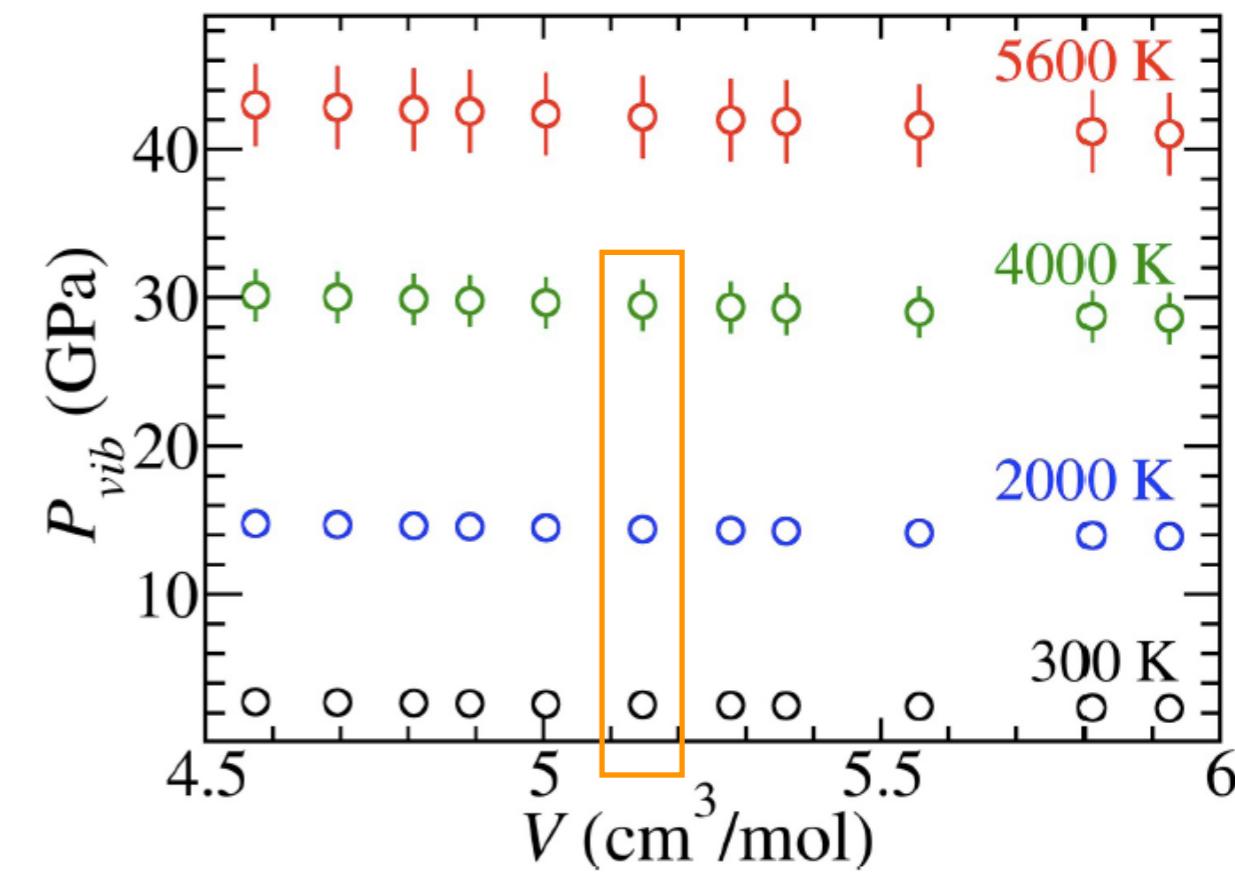
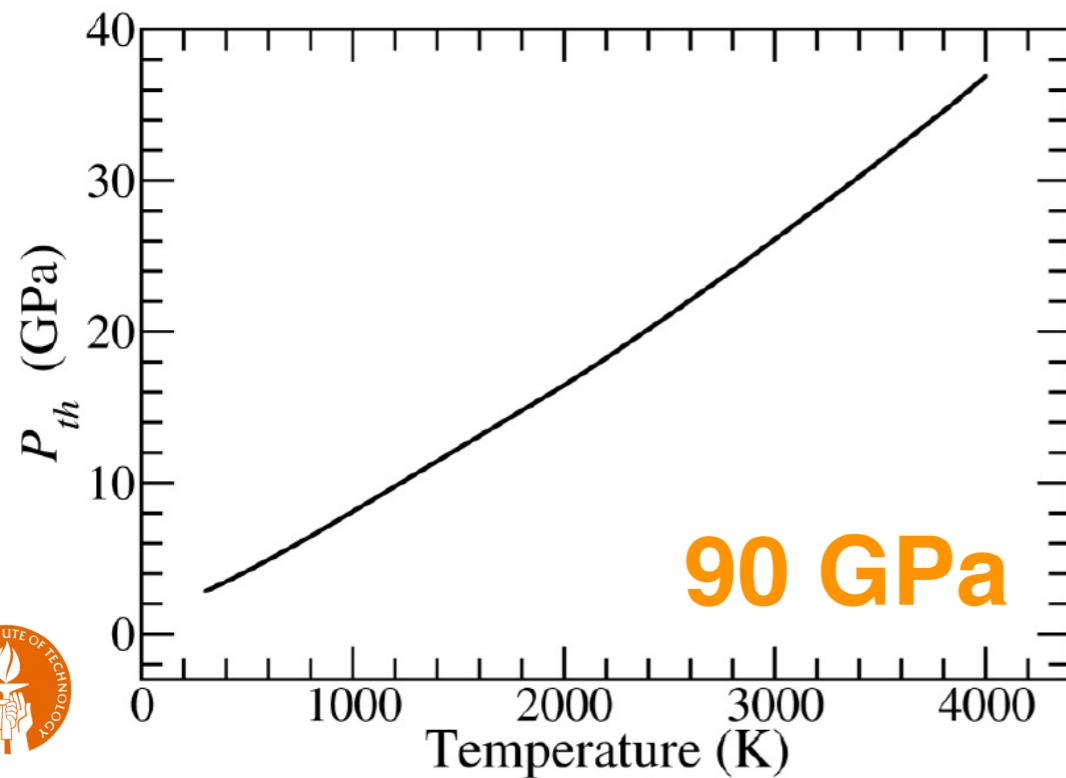
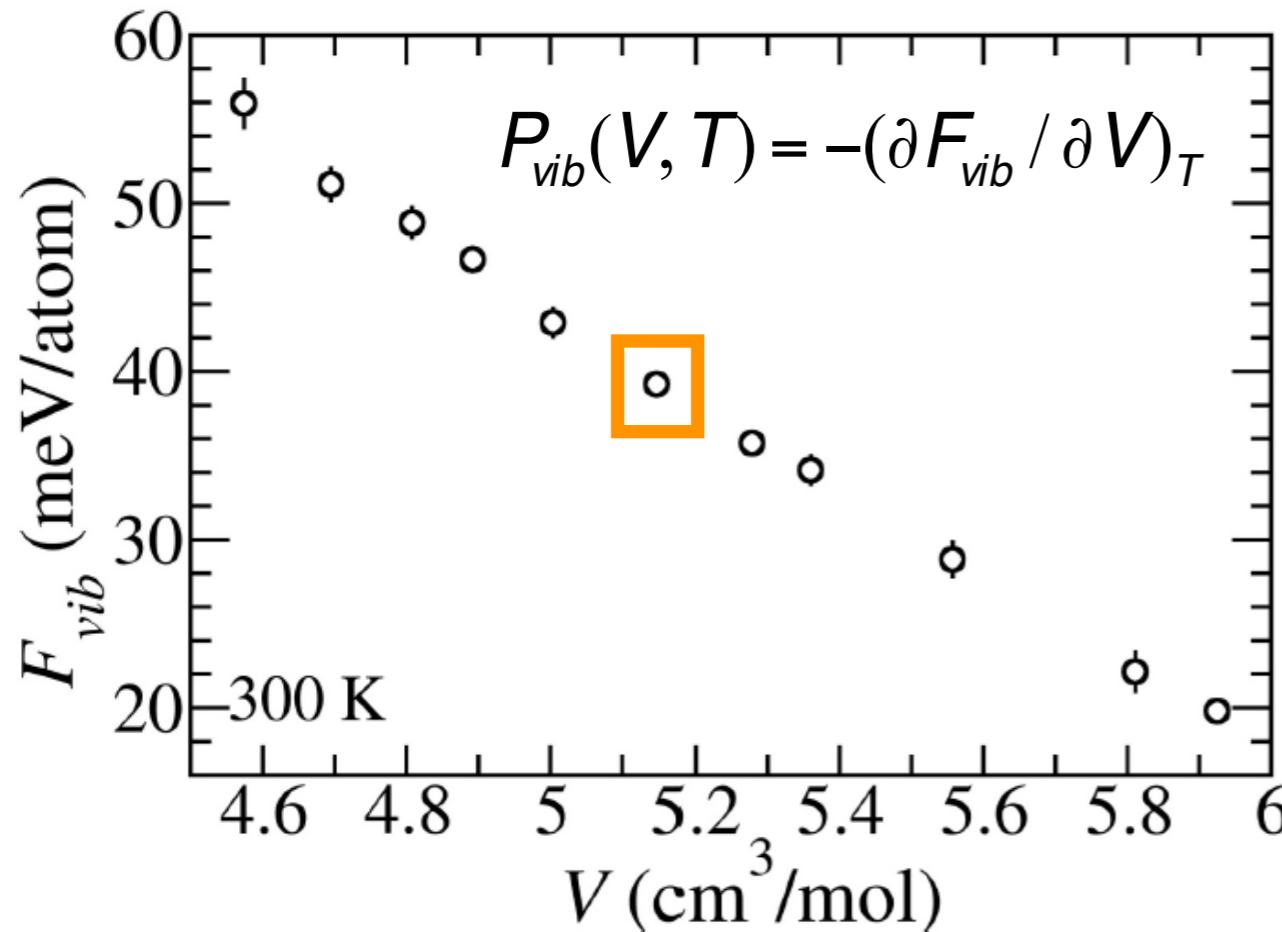
Murphy, Jackson, Sturhahn. *JGR* 2013



# Shear wave velocities of *hcp*-iron and iron-alloys from inelastic x-ray scattering measurements



# Thermal pressure from the measured volume dependence of *hcp*-iron's vibrational free energy



$$P_{th} = P_{vib}^h + P_{vib}^{anh} + P_{el}$$

Dewaele *et al.* *PRL* (2006)

Murphy *et al.* *PEPI* (2011)

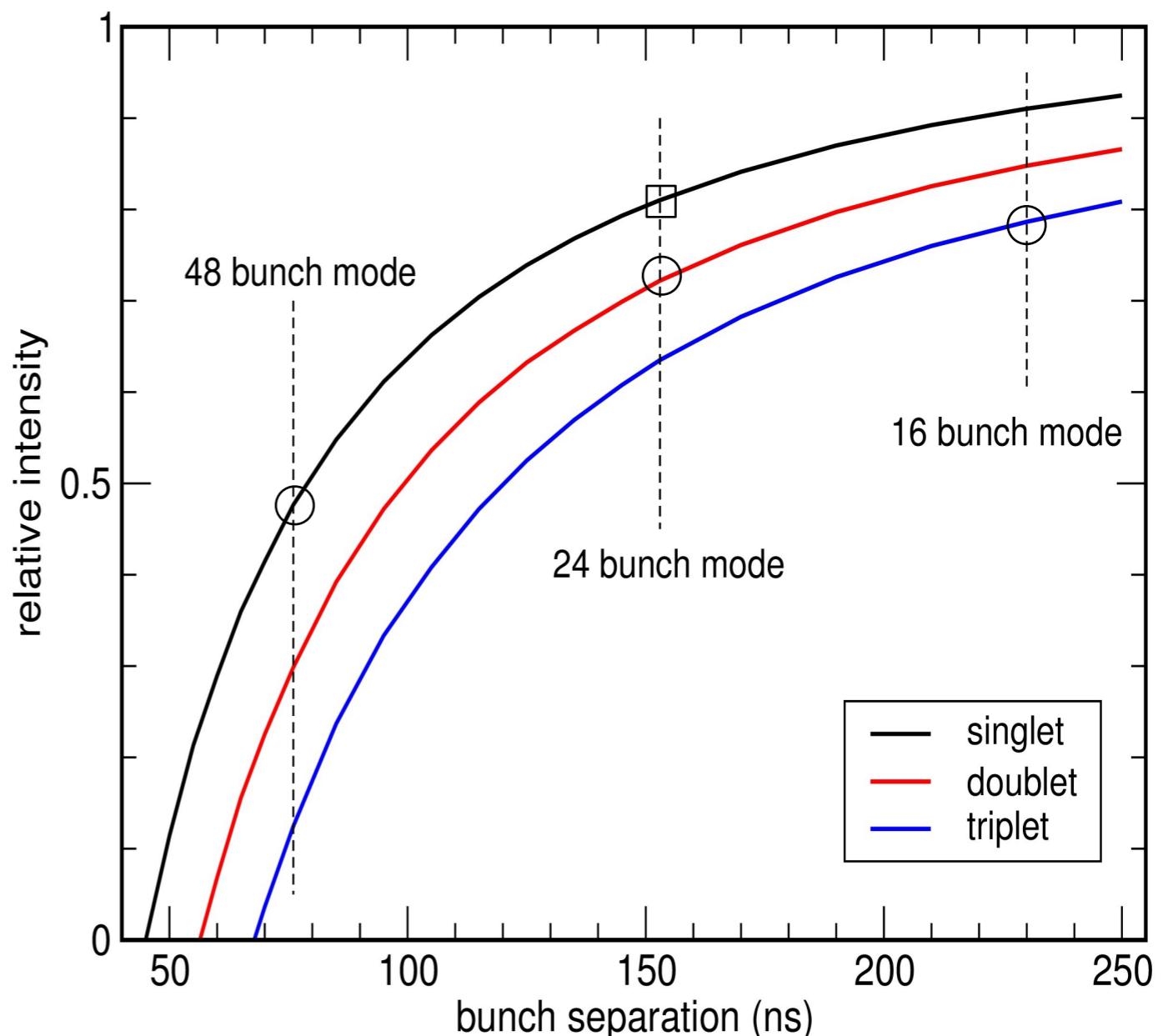
$$\rho_{ICB}^{hcp-Fe} = 13.50 \pm 0.03 \text{ g/cm}^3$$

$\Rightarrow 5.5 \pm 0.2\%$  *Density deficit*

# MBA for time-integrated studies

- Figure: time-integrated intensity for NRIXS studies of iron-bearing compounds under extreme conditions relative to 1-1296-1 ideal timing mode
- circles indicate potential MBA timing modes:
  - 48-27-1 at 47% (singlet mode)
  - 24-54-2 at 72% (doublet mode)
  - 16-81-3 at 84% (triplet mode)
- square indicates present day timing mode:
  - 24-54-1 at 81% (singlet mode)
- MBA timing modes 24-54-2 and 16-81-3 maintain data collection efficiency for multi-Mbar pressures.
- MBA timing modes 24-54-2 and 16-81-3 permit full exploit of decreased beam size, increased beam current, and optimized insertion devices.

calculations by:  
*Wolfgang Sturhahn (Caltech)*



# Goals and desired capabilities for an APS upgrade

Nature of the boundary layers between major seismic structures  
Interaction region between the base of the mantle and the outer core  
Inner core boundary  
Magnitude, nature, origin of inner core seismic anisotropy

Time resolved, kinetics of reactions  
Complex and chemically heterogeneous materials; nature of interfaces  
Polyamorphism in glasses, liquids  
To determine shear elasticity, melting, EOS, crystal/micro- structures, valence, spin, and magnetism of Earth materials at  $>1 \text{ TPa}$   $>10,000 \text{ K}$

**Mössbauer spectroscopy:** 4D imagining of Fe valences, Melting

**(Nuclear resonant) Inelastic x-ray scattering:** phonons, free energy, entropy, elasticity, anisotropy

$>1 \text{ TPa}$   $>10,000 \text{ K}$

**Requirements:** Time structure and purity are essential for nuclear resonant scattering: Double or triple bunch mode desired for Fe; at least 48 bunch mode with high speed shutter

Smaller source, Higher flux, Better collimation

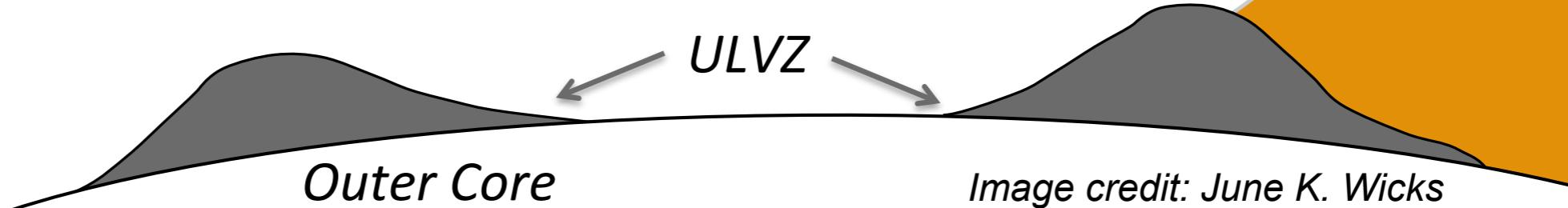


Image credit: June K. Wicks